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SUMMARY

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The reflectance and optical constants of evaporated gold films were measured in the vacuum ultraviolet from 300 Å to 2000 Å. The optical constants were determined from reflectance measurements made at various angles of incidence. In contrast to aluminum, gold shows little change in reflectance after deposition in vacuum and during exposure to air. Due to interference, semitransparent films of about 150 Å on glass show the highest reflectance at most wavelengths in the vacuum ultraviolet.

AUTHOR

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INTRODUCTION

A study of the optical properties of gold in the vacuum ultraviolet is of interest for practical and theoretical reasons. The reflectance of gold is higher than that of many other film materials for wavelengths shorter than 900 Å and exhibits almost no change during extended use and storage in air. In addition, gold films are easy to deposit in any desired thickness and have proven to be an excellent medium in which to rule diffraction gratings for use in the extreme ultraviolet.

From the theoretical point of view, measurements of the reflectance and the optical constants of gold in the vacuum ultraviolet are of interest to the solid state physicist since they furnish information on the absorption spectrum and energy band structure of the metal.

It is the purpose of this paper to present the results of recent measurements of the reflectance and optical constants of gold in the wavelength region from 300 Å to 2000 Å, and to discuss the effect of film thickness on the optical properties in this region.

EXPERIMENTAL TECHNIQUES

All evaporations were performed in an evaporator-reflectometer combination in which the reflectance of the films could be measured within seconds of the completion of their deposition. The instrument, shown in Fig. 1, is attached to a one-meter normal incidence monochromator. A detailed description of the system has been published elsewhere (1), therefore only the salient features of this instrument will be mentioned here. Basically the evaporator-reflectometer consists of two vacuum chambers separated by a barrier with a hole in it. The vapor source, in this case a tungsten boat containing gold, is located in the lower chamber, while the upper chamber contains the reflectometer. The substrate holder may be disengaged from the reflectometer mechanism and placed over the hole in the barrier between the two chambers. Thus, during an evaporation, the substrate faces the boat which is approximately 40 cm away from it. The substrate and an additional shutter serve to shield the upper chamber and its contents from the metal vapor. Immediately after the completion of an evaporation the substrate may be swung up into position for measurement.

The polished glass plates used as substrates were given a preliminary cleaning with a detergent and, just prior to the evaporation, a final cleaning with a high-voltage dc glow discharge.

The evaporation rates of gold were varied so that the deposition rates ranged from 2 to 1600 Å/sec; the thickness was monitored by measuring the light transmittance at $\lambda = 5000$ Å. During the evaporations the pressure was approximately 10^{-6} torr, except when it was deliberately increased to study the effect of pressure on the optical properties.

Two types of light sources were used to cover the spectral range from 300 Å to 2000 Å. One was a dc glow discharge (1) and the other a pulsed discharge source (2). Figure 2 shows the spectra of the radiation available for measurement with the two light sources. The radiation emitted from the glow discharge is shown at the top of Fig. 2. The many-line hydrogen molecular spectrum, which within the limits of resolution of the monochromator is a quasi-continuum, and two atomic lines at 1216 Å and 1026 Å, cover the spectral region from approximately 900 Å to beyond the upper wavelength limit used in this investigation. The neutral resonance lines of He and Ne produced isolated lines at 584 Å and 736 Å, respectively, when those gases were introduced into the light source. More complete coverage of the region below 900 Å is afforded by using the pulsed discharge source. The remainder of Fig. 2 shows the radiation available using the pulsed discharge with different gases. The spectral range down to 300 Å is covered rather fully using the four gases shown. With oxygen, wavelengths as short as 246 Å may be obtained.

The reflectance of the films could be measured at any angle of incidence between 6^{o} and 86^{o} using a 1P21 photomultiplier sensitized with sodium salycilate as a detector. For the determination of optical constants by the reflectance method described by Tousey (3) and Simon (4), reflectance measurements were made at several angles of incidence at each wavelength. The resulting values of reflectance were then used to determine the possible values of the index of refraction n and of the extinction coefficient k (the complex index of refraction, $\mathfrak{N} = n - ik$) for each angle of incidence by means of previously computed curves. The values of n and k common to each determination for several angles of incidence then gave the optical constants at the wavelength under study. These initial values were then fed into a digital computer which was programmed to calculate the reflectance for each angle at which measurements were made and to evaluate the deviation between the measured and calculated values. The constants were then varied by small amounts and the calculation repeated until the deviation was minimized. The program also computed the reflectance at normal incidence which, in all cases, agreed within experimental error with the measured values.

RESULTS

Evaporated gold shows little change in vacuum ultraviolet reflectance after its deposition in vacuum, and during exposure to and prolonged storage in air. The marked contrast in behavior to that of a reactive metal such as aluminum may be seen in Fig. 3. In separate experiments, the reflectances of freshly deposited aluminum and gold were studied as a function of time at 1216 Å, before and after exposure to air. The reflectance of aluminum drops rapidly, particularly upon exposure to air, due to the formation of a strongly absorbing oxide layer (5). The reflectance of gold, however, decreases only about one percent while in vacuum and stays essentially constant after exposure to air. At other wavelengths the degree of change was about the same, but at some wavelengths a slight increase could be observed. Contamination resulting from storage in air sometimes caused a slight change in reflectance at all wavelengths, however, a simple rinse with ethyl alcohol restored the reflectance practically to its original value. Thus the conclusion was reached that measurements *in situ* were unnecessary, and that the true optical properties of gold could be studied using films that had been exposed to air.

The effect of deposition parameters on the reflectance of gold films was investigated, and it was found that neither the pressure during deposition nor the rate of deposition were critical until extremes were reached. Deposition rates of from 50 to 1600 Å per second with pressures of from less than 10^{-6} to above 10^{-5} torr were employed with practically no effect on the reflectance of the resulting film. Only when a rather thick film was made under poor conditions did the reflectance drop, undoubtedly due to increased surface roughness. In Fig. 4 the upper solid curve represents the reflectance of a film made in a more or less typical evaporation; the lower is the reflectance of a film made under the relatively poor conditions given. The reflectance values of Robin (6), obtained from visibly opaque films and shown as the dashed curve of Fig. 4, are considerably lower than those measured here.

If the surface of the evaporated gold film is rough, it will scatter the incident radiation and cause erroneous reflectance measurements which will result in inaccurate determinations of n and k. Since roughness increases with increasing film thickness, the films should be kept thin to avoid roughness but, at the same time, thick enough to be opaque. Interference effects and transmission losses will be negligible if less than 0.1% of the energy incident on the vacuum-film interface is transmitted through the film to the glass substrate. Calculations of the film thickness required to satisfy this condition were made using preliminary values for n and k obtained from a film 1600 Å thick. A plot of the thickness necessary to reduce transmission to 0.1% as a function of wavelength from 300 Å to 2000 Å is shown in Fig. 5. The optical constants given by Schulz (7) and Schulz and Tangherlini (8) were used to calculate the transmittance of gold at 5000 Å as a function of thickness, and the right-hand scale of Fig. 5 gives the transmittance at 5000 Å corresponding to the thickness given in the left-hand scale. It is seen that films that are opaque in the vacuum ultraviolet are semitransparent at 5000 Å. Thus monochromatic light of that wavelength may be used to monitor the thickness of gold films that will be opaque in the vacuum ultraviolet.

Once the desirable thickness for gold films was determined for the various wavelengths, measurements of n and k were made using films of the appropriate thicknesses. The results are shown in Fig. 6, where n, k, and the measured normal incidence reflectance are plotted as a function of wavelength from 300 Å to 2000 Å. Cole and Oppenheimer (9) have measured the optical constants of gold at the wavelengths 1216, 1048, 920, 584, and 304 Å. Their values of n and k are, with the exception of the indices at 584 Å and 304 Å, lower than the ones reported here.

The extinction coefficient, k, cannot be correlated directly with absorption processes occurring in the metal, however, Frohlich and Pelzer (10) have shown that a phenomenological relation exists between absorption processes and the bulk optical constants. According to them, the quantity,

$$X = 2nk/(n^2 + k^2)^2 ,$$

when plotted against wavelength, should have maxima corresponding to absorption processes.

This expression was calculated using the n and k values of Fig. 6 and the results are shown in Fig. 7. There are distinct maxima; at 32.6 ev, 25.8 ev, 16.3 ev, and 6.7 ev, corresponding to wavelengths 380, 480, 760, and 1880 Å.

Robins (11) used a reflection technique to study mono-energetic electrons elastically and inelastically scattered by an evaporated gold surface and observed four characteristic losses of about the same energy; 32.6 ev, 25.8 ev, 16.0 ev, and 6.3 ev.

Interference effects may be observed when very thin gold films are deposited on glass. Figure 8 shows the calculated and measured normal incidence reflectances of opaque and 150 Å thick films from 1000 Å to 2000 Å. The agreement between calculated and measured values for the 150 Å thick film indicates that n and k of the bulk material are still correct for films this thin. The curve also shows that highest reflectance is obtained with films that are not opaque in the extreme ultraviolet. The same situation applies to films of platinum (12).

The calculated reflectance of evaporated gold on glass as a function of film thickness at several wavelengths is shown in Fig. 9. At all wavelengths shown except 700 Å there is an increase in reflectance at a gold thickness less than about 250 Å. It can be concluded that for use as a reflector throughout the vacuum ultraviolet a gold thickness of about 150 Å on glass represents a good compromise.

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LIST OF CAPTIONS

Figure 1. Section of evaporator-reflectometer combination.

Figure 2. Spectra produced by the two light sources.

Figure 3. Comparison of the effect of aging on the reflectance of evaporated films of gold and of aluminum at 1216 Å.

Figure 4. Effect of evaporation conditions on the reflectance of evaporated gold as a function of wavelength from 1000 Å to 2000 Å.

Figure 5. Calculated thickness of gold films required to decrease transmittance to 0.1% as a function of wavelength from 300 Å to 2000 Å. Transmittance data at 5000 Å are included for comparison.

Figure 6. Optical constants and normal incidence reflectance of gold in the vacuum ultraviolet.

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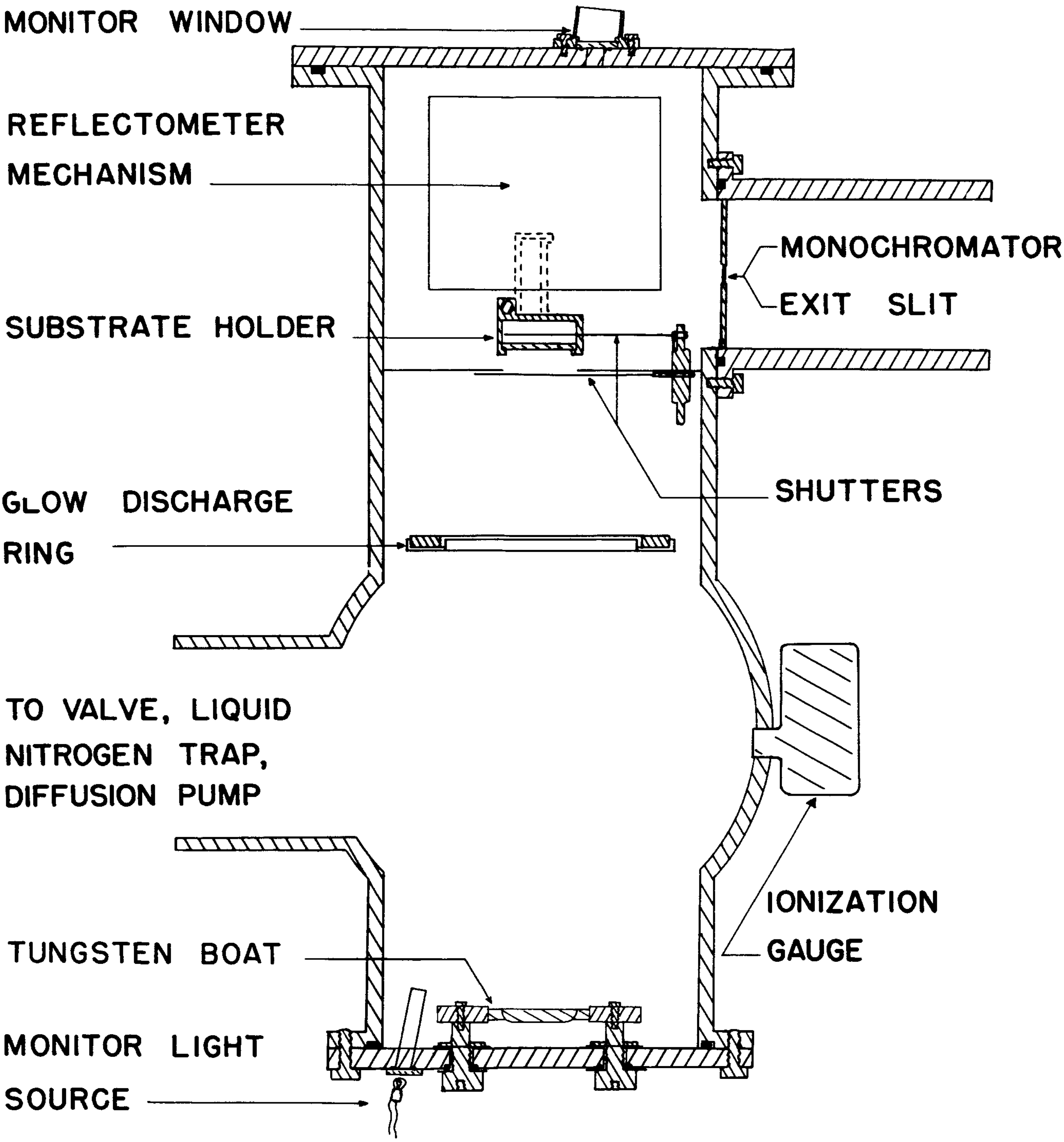
Figure 8. Calculated and measured reflectance of opaque and semi-transparent ($t = 150$ Å) gold films on glass as a function of wavelength from 1000 Å to 2000 Å.

Figure 9. Calculated reflectance of gold on glass as a function of film thickness for various wavelengths in the vacuum ultraviolet.



SECTION OF EVAPORATOR - REFLECTOMETER COMBINATION

Figure 1

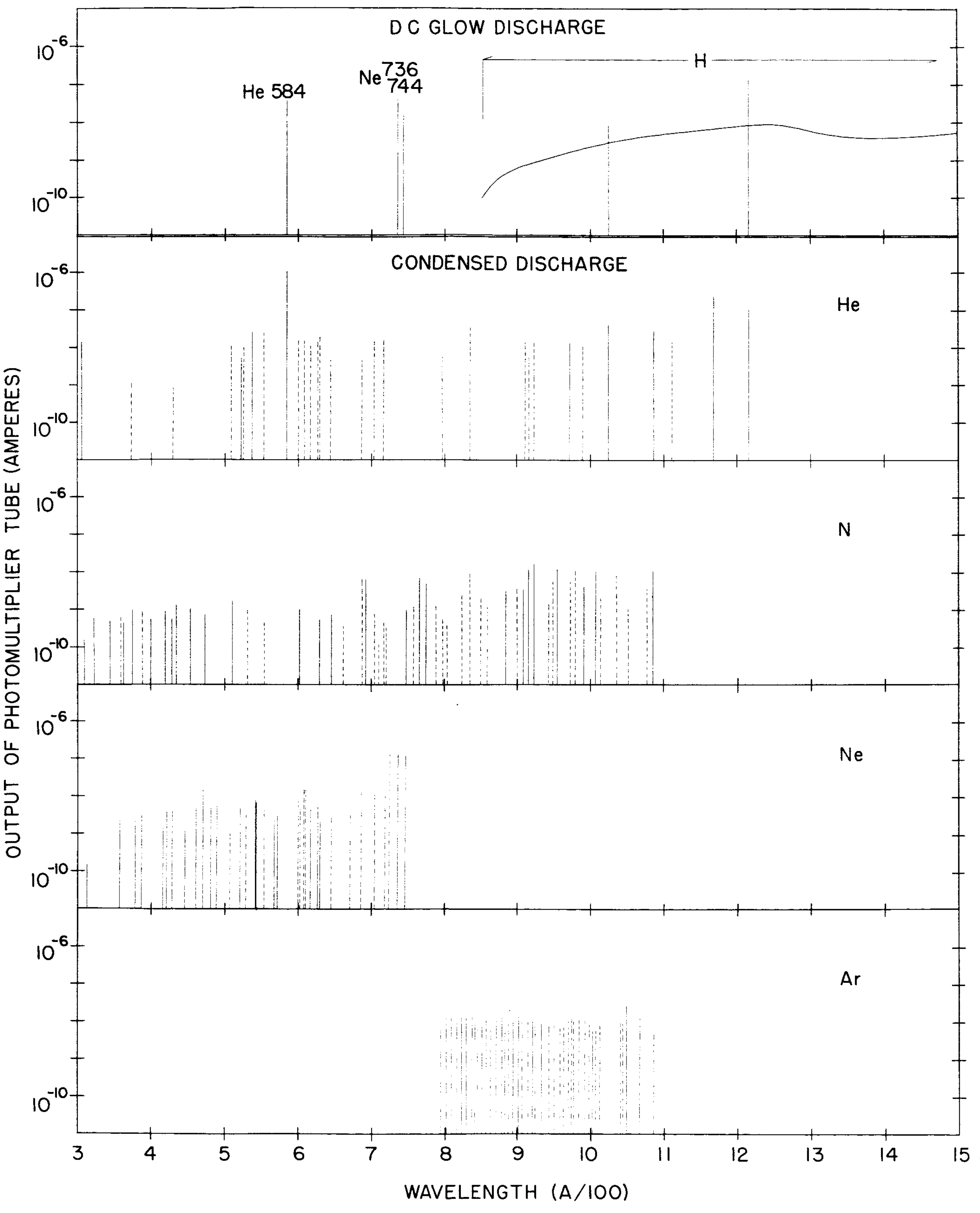
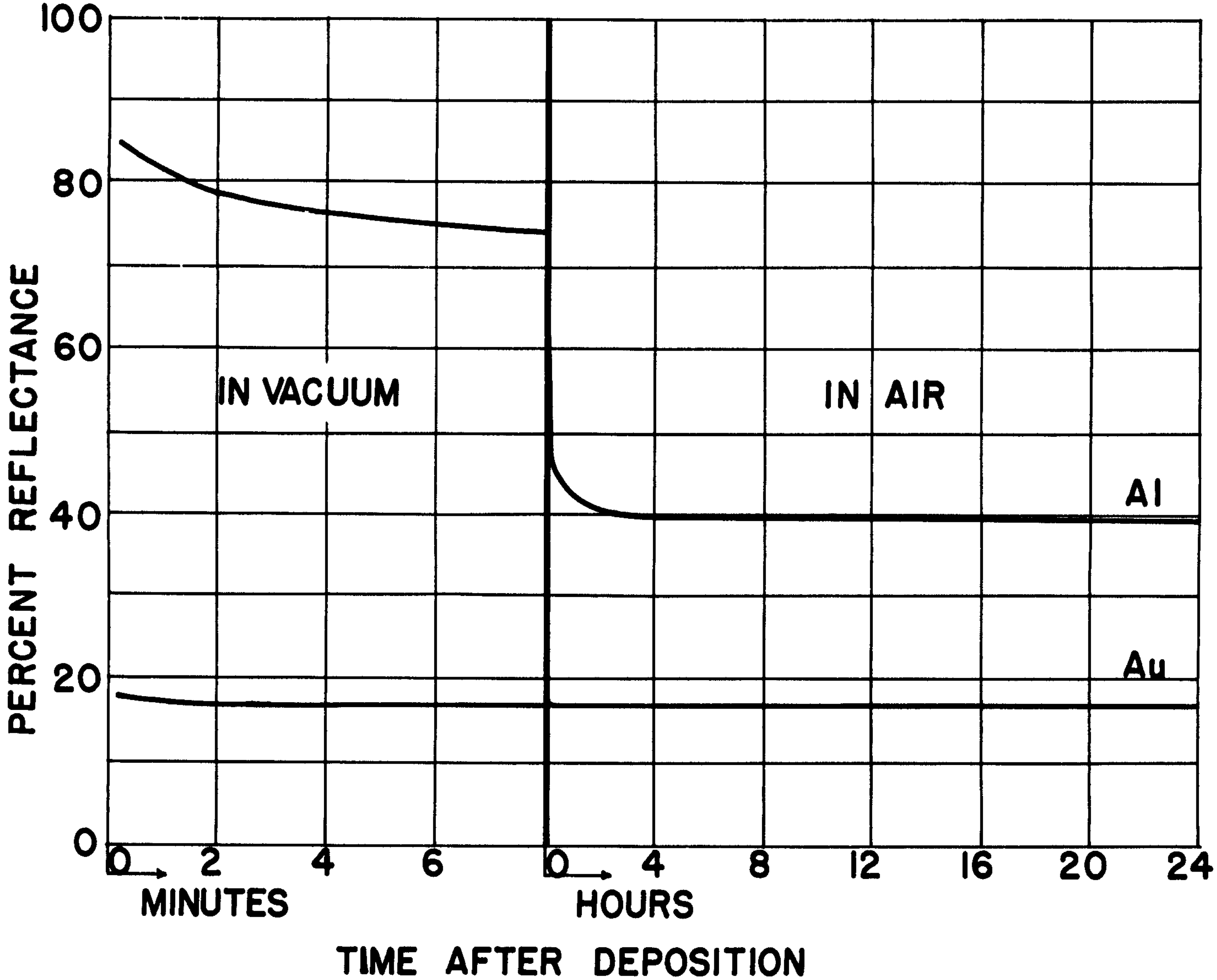
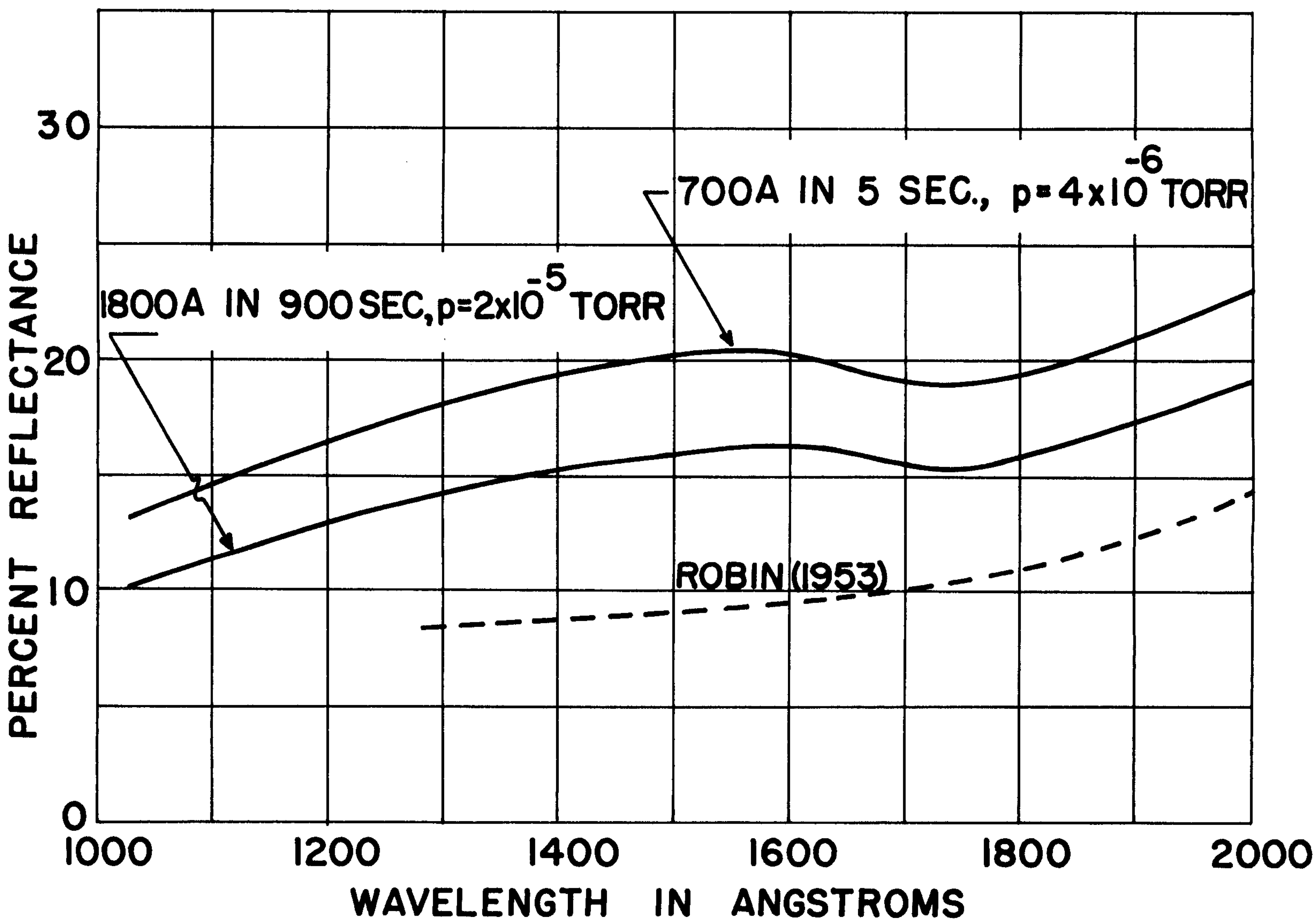

DC GLOW DISCHARGE
He 584
Ne 736 744
H
CONDENSED DISCHARGE
He
N
Ne
Ar
OUTPUT OF PHOTOMULTIPLIER TUBE (AMPERES)
WAVELENGTH (A/100)

Figure 2



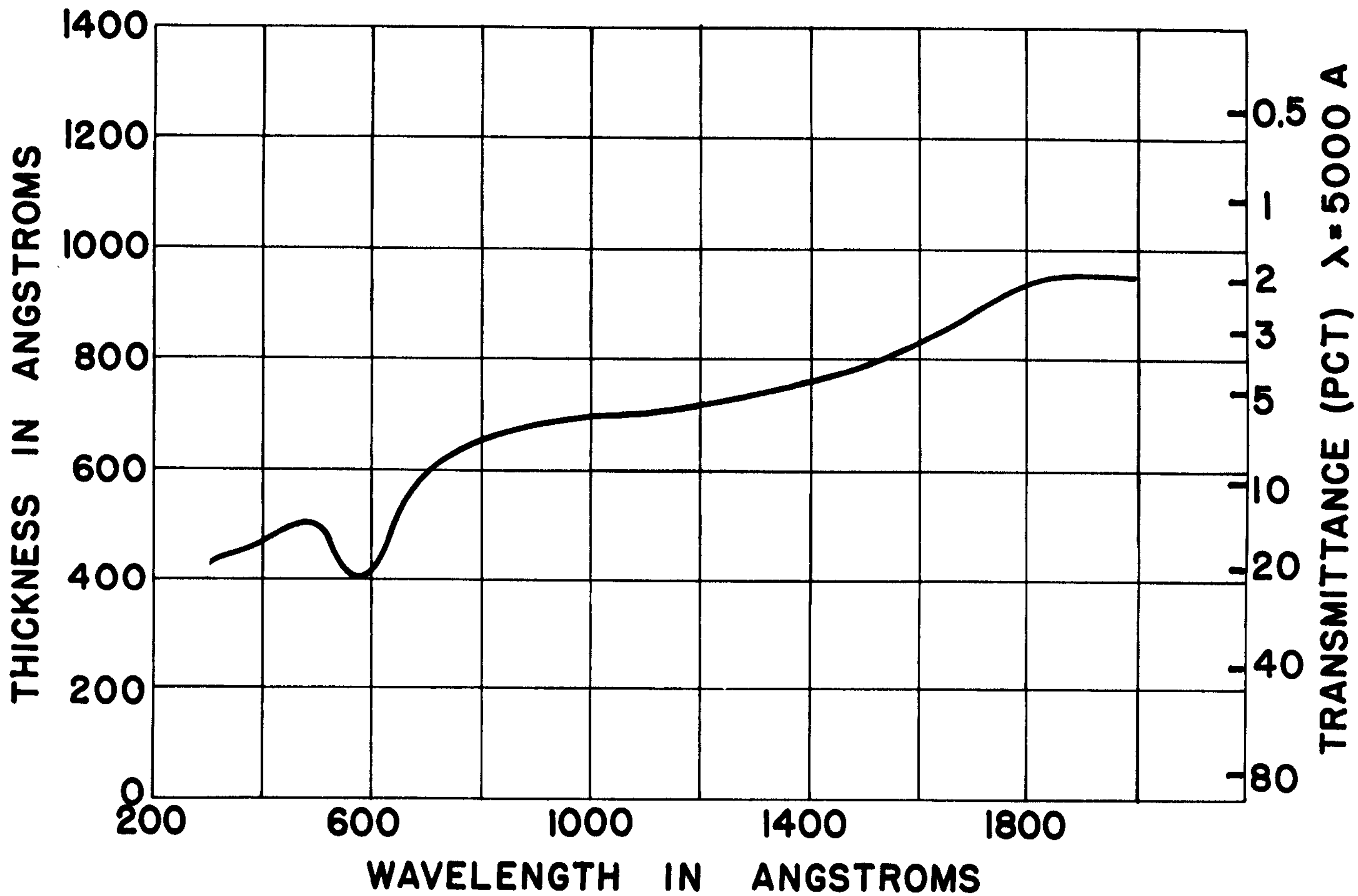
COMPARISON OF THE EFFECT OF AGING ON THE REFLECTANCE OF EVAPORATED FILMS OF GOLD AND OF ALUMINUM AT λ1216A.

Figure 3



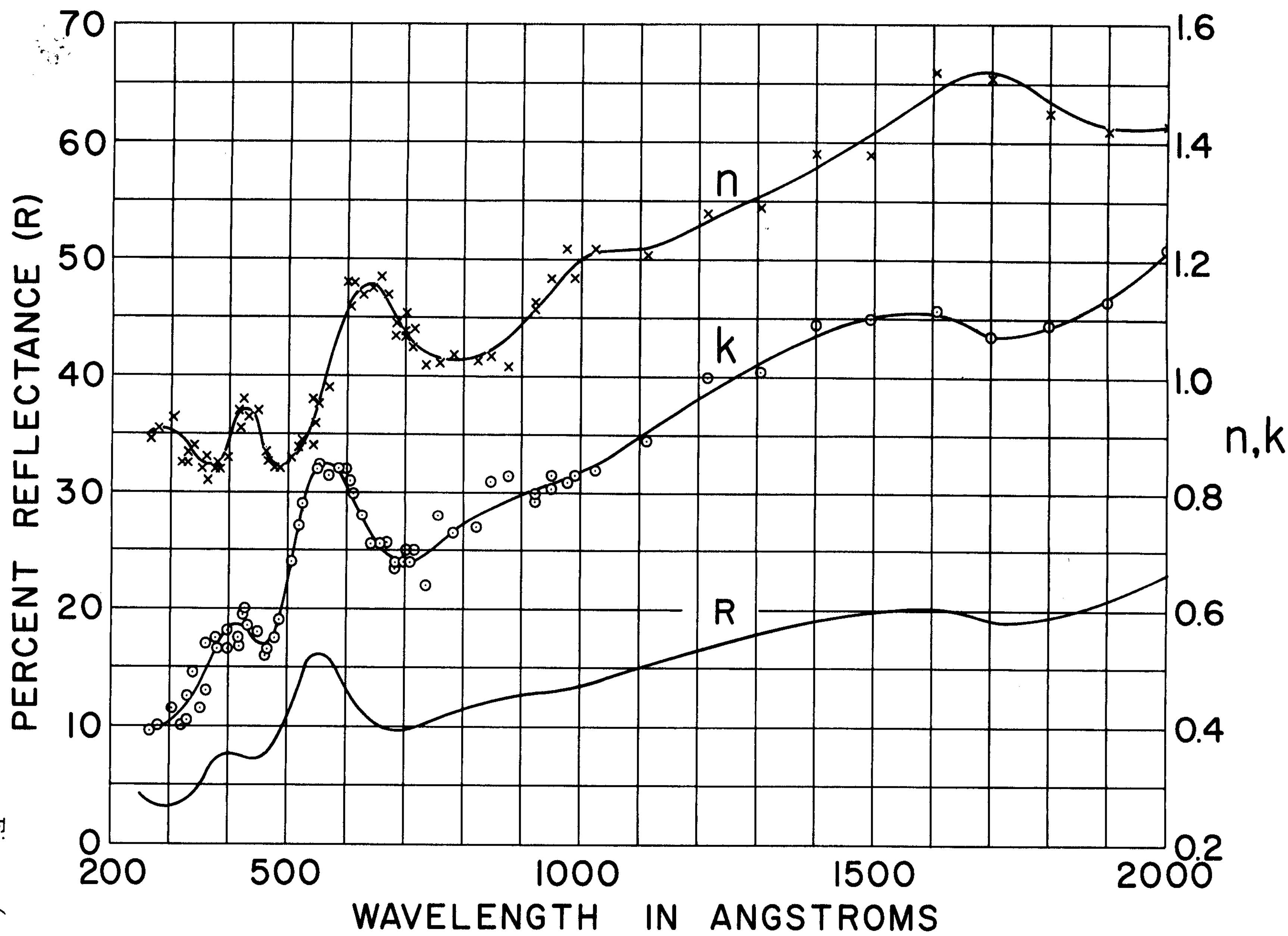
EFFECT OF EVAPORATION CONDITIONS ON THE REFLECTANCE OF EVAPORATED GOLD AS A FUNCTION OF WAVELENGTH FROM λ1000A TO λ2000A.

Figure 4



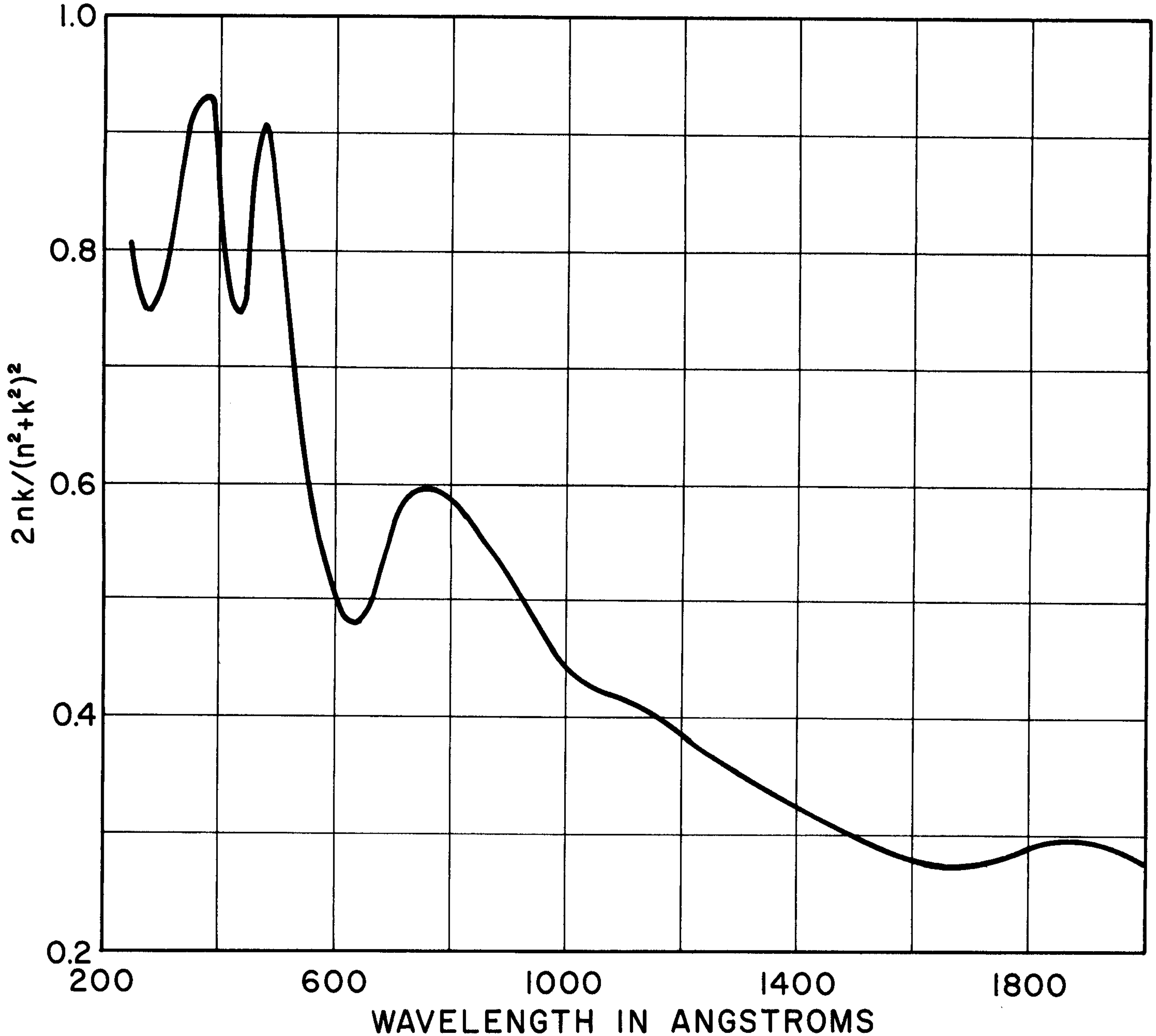
CALCULATED THICKNESS OF GOLD FILMS REQUIRED TO DECREASE TRANSMITTANCE TO 0.1% AS A FUNCTION OF WAVELENGTH FROM λ300A TO λ2000A. TRANSMITTANCE DATA AT λ5000A ARE INCLUDED FOR COMPARISON.

Figure 5



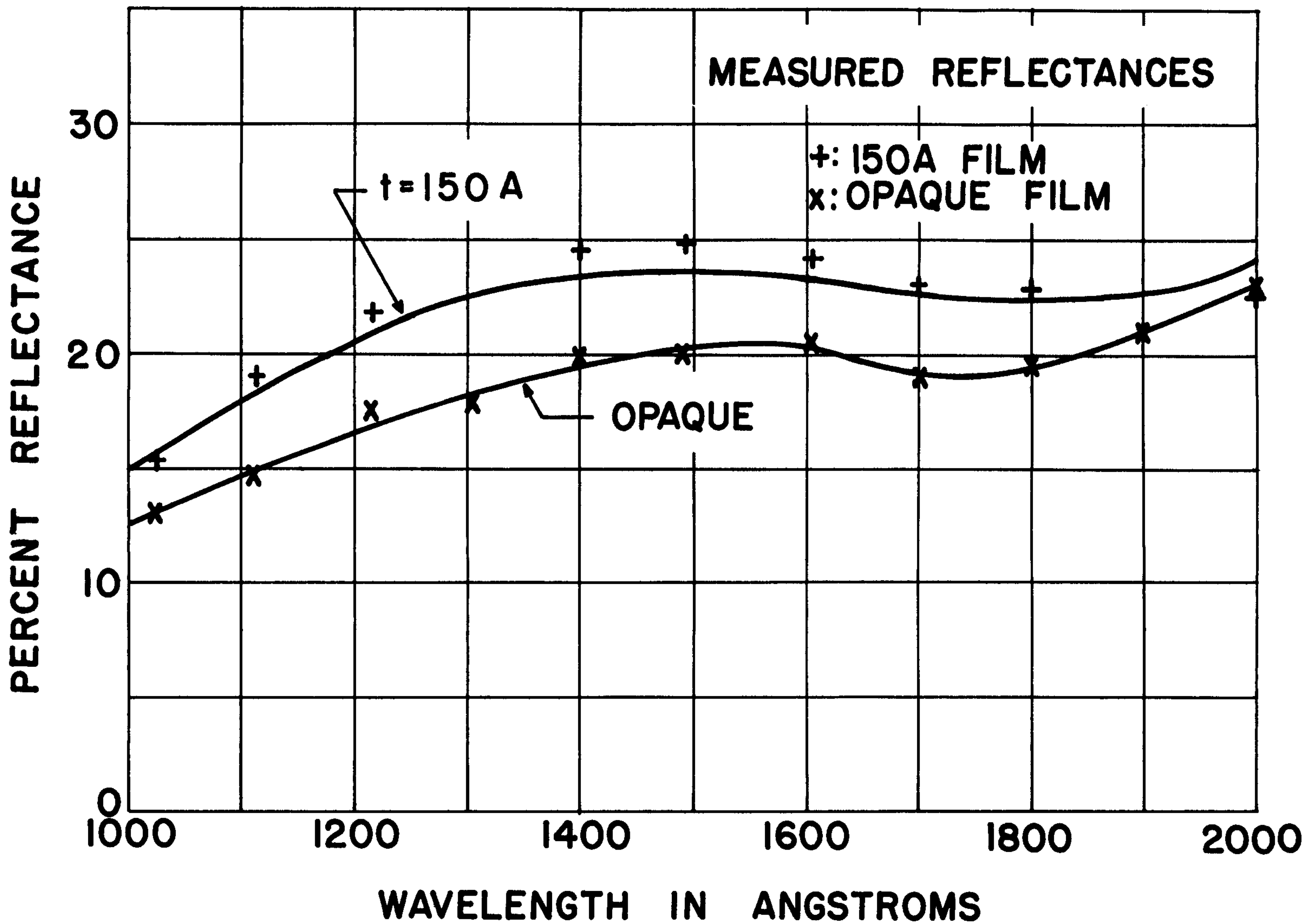
OPTICAL CONSTANTS & NORMAL INCIDENCE REFLECTANCE OF GOLD IN THE EXTREME ULTRAVIOLET.

Figure 6



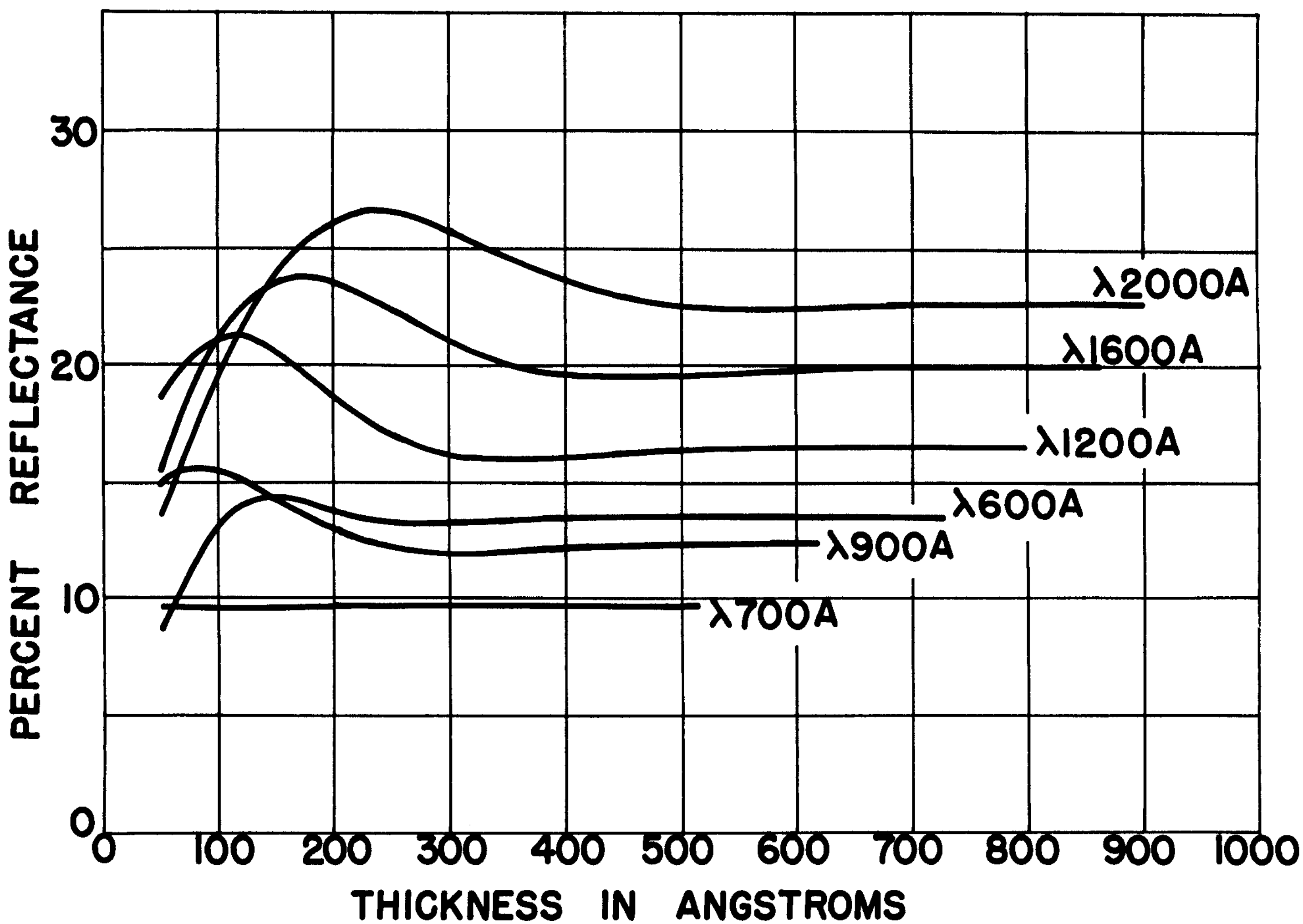
$2nk/(n^2+k^2)^2$ AS A FUNCTION OF WAVELENGTH FOR GOLD.

Figure 7



CALCULATED AND MEASURED REFLECTANCE OF OPAQUE AND SEMITRANSPARENT (t=150 A) GOLD FILMS ON GLASS AS A FUNCTION OF WAVELENGTH FROM λ1000A TO λ2000A.

Figure 8



CALCULATED REFLECTANCE OF GOLD ON GLASS AS A FUNCTION OF FILM THICKNESS FOR VARIOUS WAVELENGTHS IN THE VACUUM ULTRAVIOLET.

Figure 9